

## Neutron Capture Experiments -Filling Nuclear Data Needs for Safeguards

B. W. Sleaford

August 3, 2015

INTERNATIONAL NUCLEAR MATERIALS MANAGEMENT Indian Wells, CA, United States
July 12, 2015 through July 16, 2015

#### Disclaimer

This document was prepared as an account of work sponsored by an agency of the United States government. Neither the United States government nor Lawrence Livermore National Security, LLC, nor any of their employees makes any warranty, expressed or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States government or Lawrence Livermore National Security, LLC. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States government or Lawrence Livermore National Security, LLC, and shall not be used for advertising or product endorsement purposes.

# Neutron Capture Experiments – Filling Nuclear Data Needs for Safeguards

Non Destructive Assay Working Group Meeting

**International Nuclear Materials Management** 

**Brad Sleaford** 

July 12, 2015

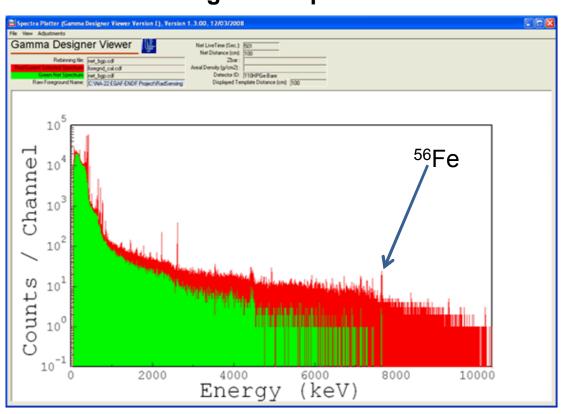


### LLNL-CONF-675653

This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under contract DE-AC52-07NA27344. Lawrence Livermore National Security, LLC

- -Nuclear Data upgrades for Transport Modeling
- -Neutron Capture gamma spectroscopy,
- -HPGe detectors

### Measurement of gamma spectra from Pu mass-> Want to Model This



### **Capture Gammas:**

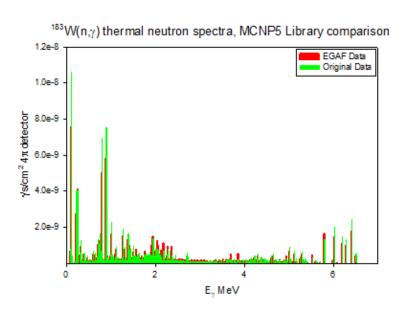
- -High Energies to 12 MeV
- -Fingerprint of Isotope
- -Neutrons=Actinides

Used in ANY spectroscopy application, Emergency response, Safeguards, etc.

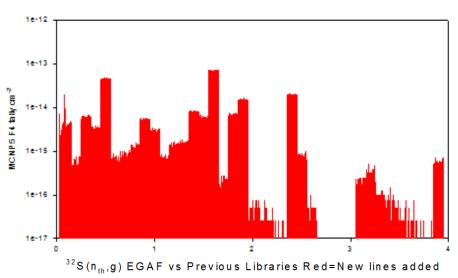




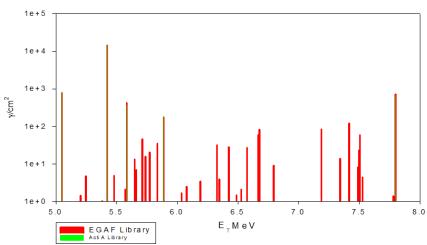
# **Examples of Missing Capture Data and some improvements for Transport Applications**



Some libraries have no spectra P, (Actinides)
Others have poor resolution, missing lines, etc.

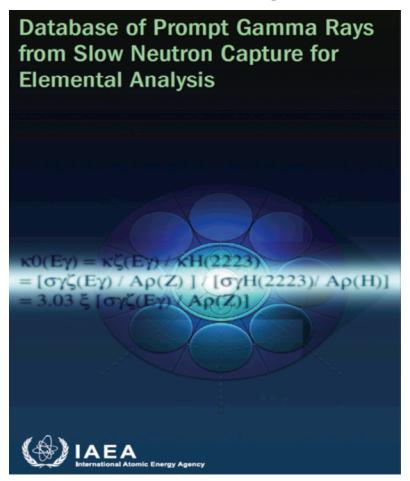


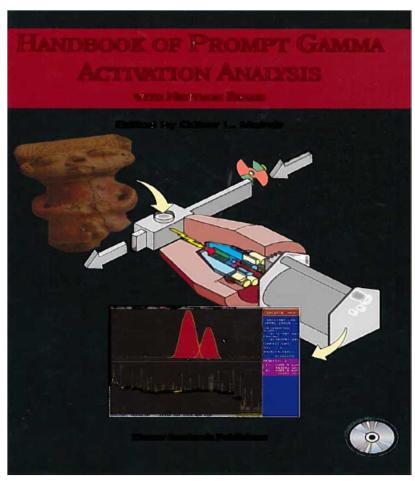
Pb ENDF/B VII (n,γ) spectra





# IAEA CRP Capture Gamma Spectra for ~260 Nuclei 36K lines ~ Budapest Reactor Measurements + FRMII





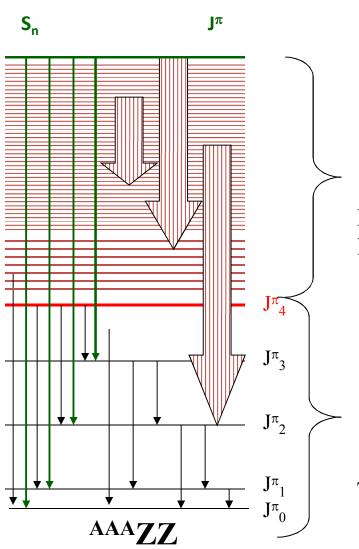
Rick Firestone, Tamas Belgya, Zsolt Revay, et. al.

## **Evaluated Gamma-Ray Activation File (EGAF)**

-Cross correlated with ENSDF decay schemes MCN P5 runs: Pd104 + n<sub>thermal</sub> Origional ENDF library vs EGAF library -Put into ENDF libraries for transport modeling 25e-9 to be in next ENDF release and 1st libraries. Origional Library EGAF Library 20e-9 available through BNL now v/cm² into 4∏ PGAA Beta Prompt 1.0e-9 **Particle** Gamma ray Target 5.0e-10-Nucleus Incident Neutro E\_MeV (5 KeV bin widths~ HPGe resolution) Radioacti Nucleus Figure 2 **Budapest Reactor** Product neutron beam **DICEBOX** Nucleus Fission products Delayed Compound statistical Gamma ray NAA **Nucleus** model code Evaluated **Reaction Input** Nuclear **ENSDF** Parameter Library **Evaluated Gamma-ray Data File** database (RIPL) Activation File (EGAF) (ENDF)



## Gamma Cascade Quasi-Continuum in Medium-Heavy Nuclei modeled with Extreme Statistical model Dicebox (F. Becvar, M. Krticka)



Thermal neutron capture state energy and  $J^{\pi}$  value(s) are taken from experiment if known.

Monte-Carlo cascade: Capture->Ground State
Continuum=Energy bins above critical energy
Partial widths calculated from presumed known
level density and Strength function models

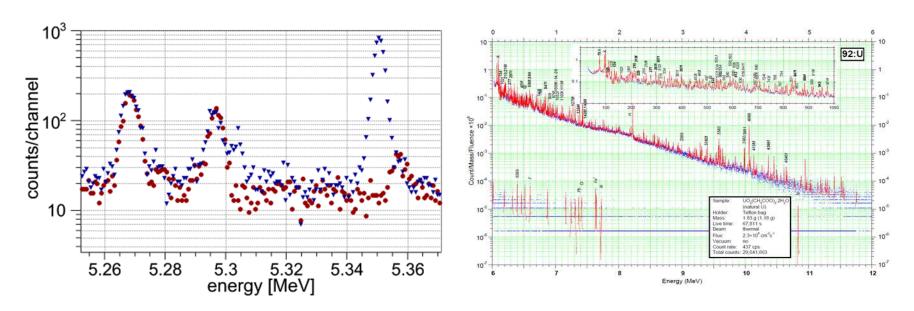
EGAF Data experimentally measured below critical energy

Total feeding to ground state now available= $\sigma_{total}$ 



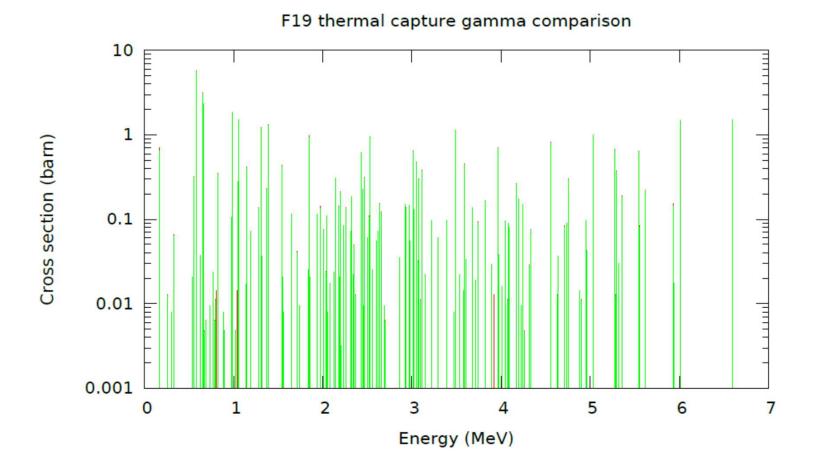
# Thermal Capture Gammas from Actinides New experimental Data being evaluated

Probable Primary Np237 line at 5.35 MeV U235 Capture line @ 6.395 MeV is 4 mb



There are no high resolution capture gamma lines in the major actinides in ENDF

## F had few changes



## 26 beta Libraries in testing->2 to BNL (Feb 2015)

Z		A	%NA	barns	number of Gammas
1	Н	1	99.9844	0.33260	1
1	D	2	0.01557	0.00052	1
3	Li	6	7.589	0.04000	3
3	Li	7	92.411	0.05000	3
4	Be	9	100	0.01000	12
5	В	10	19.82	0.50000	9
5	В	11	80.18	0.01000	9
6	C	12	98.892	0.00353	6
7	N	14	99.6337	0.08000	60
8	0	16	99.7628	0.00019	4
9	F	19	100	0.00960	165
11	Na	23	100	0.53000	233
12	Mg	24		0.06000	283
13	Al	27	100	0.23000	291
14	Si	28	92.2297	0.18000	54
15	P	31	100	0.17000	202
16	S	32		0.53000	470
17	Cl	35	75.771	45.55000	383
17	Cl	37	24.229	0.43000	77
26	Fe	56	91.75	2.59000	193
46	Pd	104	11.14	0.60000	13
74	W	182	26.4985	19.90000	126
74	W	183	14.3136	10.30000	212
74	W	184	30.6422	1.70000	64
74	W	186	28.4259	38.50000	152
82	Pb	207	22.0827	0.62500	25





### **Capture Gamma Applications-Active Interrogation**



Evaluating munitions for presence of explosives, chemical or nerve agents

Idaho National Lab-80 units in use worldwide

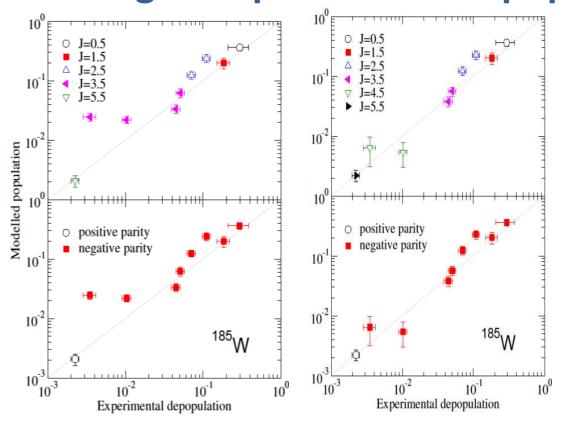
Spontaneous Fission, DD and DT neutron sources used

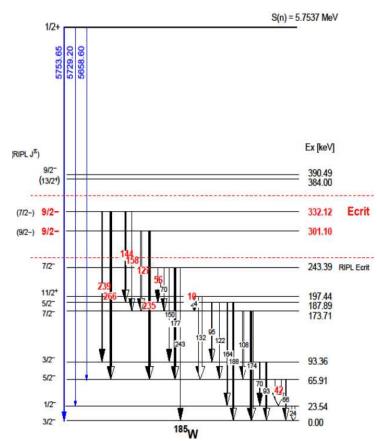
Gus Caffrey, Ed Seabury, INL



... \_ \_ \_ \_ \_ \_ \_ \_ .

# IAEA/EGAF targets are Elemental-ENDF libraries are Isotopic: Tungsten ENSDF Data had inaccuracies, <sup>185</sup>W, 9 levels below Ecrit— Dicebox population feeding vs Experimental depopulation





Aaron Hurst, et. al.





### **Neutron Spectra from Passive and Active Sources:**

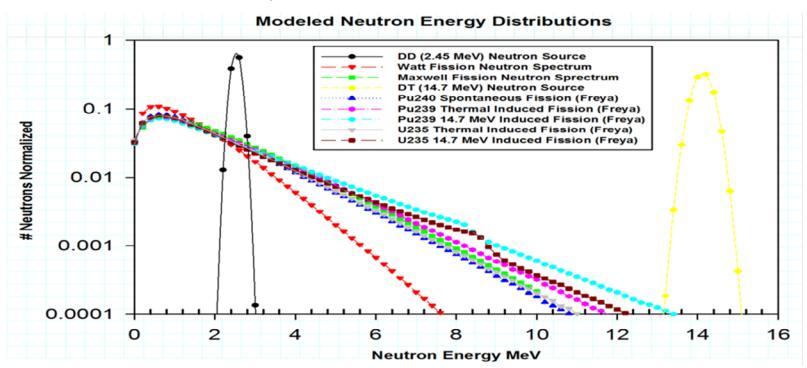
IAEA data is thermal reactor neutrons-Capture Dominates Passive counting is mostly Spontaneous Fission neutrons Active interrogation:

DD spectra 2.45 MeV

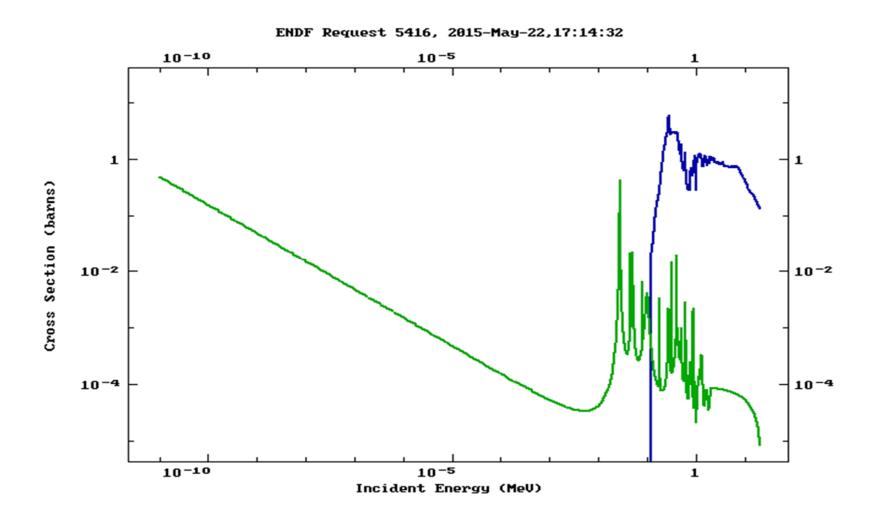
DT spectra 14 MeV

National Ignition Facility is special case

Neutrons thermalize on ~μs time scale



### **At Higher Neutron Energies Inelastic lines dominate**



# A Carefully Done 1978 Atlas of HPGe Spectra from ~ 700 keV Neutrons: another Useful Database?

## ATLAS

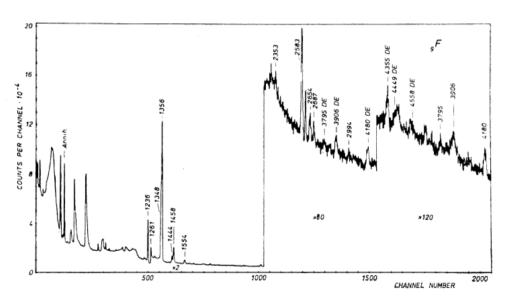
# OF GAMMA-RAY SPECTRA FROM THE INELASTIC SCATTERING OF REACTOR FAST NEUTRONS

#### Level scheme of 19F [72Aj]

$E_{\vec{L}}$	$E_{i}^{a}$	$J_{\hat{i}}^{\pi}$	Ε <sub>γ</sub>	· I <sub>7</sub>	Ef	J <sub>f</sub> <sup>∞</sup>	$P_S$
109.9 197.1 (2)	109.893 197.147	1/2- 5/2+ 5/2-	197.1	2700		1/2+	2390
1345.74 (10) 1458.4 (4)	1345.72 1458.5	5/2- 3/2-	1235.80 1458.4	100 33	109.9 0	1/2-	91 173
.,		,	1348.0 1261.1	120 20	109.9 197.1	1/2- 5/2+	
1553.9 (5)	1554.1	3/2+	1554.0 1444.0	8.0 15	0 109.9	1/2+	2 <b>8</b> 6
2779.9 (3)	1779.80 3907.1	9/2+ 3/2(+)	1356.5 2582.6 3905.9	$   \begin{array}{r}     265 \\     20 \\     \hline     3.5   \end{array} $	197.1 197.1 0	5/2+ 5/2+ 1/2+	20 6.5*
3906.2 (15)	3907.1	3/2(+)	3794.9 2352.6	1.0 1.0	109.9 1553.9	1/2- 3/2+	0.5
3999.6 (6) 4032.5 (6)	399 <b>8.5</b> 4032 <b>.</b> 5	7/2- 9/2-	2653.8 2686.6	5.4 3.7	1345.7 1345.7	5/2- 5/2-	6.8* 3.7
4377.9 (16) 4552.4 (225	4377.7 4555	7/2+ 5/2+	4180.2 4354.8	3.0 2.0	197.1 197.1	5/2+ 5/2+	4.0* 3.0*
4558.9 (5))	4557.5	3/2-	4558.3 4449.3	$\frac{2.0}{2.0}$	0 109.9	1/2+	4.9

A. Demidov, et. al. Kurchatov Institute, Moscow

## <sup>19</sup>F Spectra Atlas 2 ENDF?



9**F** 

### Atlas (n,n')

### ENDF (n,n')

Fluorine		MF3 MT51	1.099000+5
<u>197.1</u>	<u>8.17E-01</u>	<u>52</u>	<u>1.970000+5</u>
1235.8	3.03E-02		
1261.1	6.05E-03		
<u>1348</u>	3.63E-02	<u>53</u>	<u>-1.346000+6</u>
1356.5	8.02E-02		
1444	4.54E-03		
<u>1458.4</u>	<u>9.98E-03</u>	<u>54</u>	-1.459000+6
<u>1554</u>	2.42E-03	<u>55</u>	-1.554000+6
2352.6	3.03E-04	56	-2.780000+6
2582.6	6.05E-03		
2653.8	1.63E-03		
2686.6	1.12E-03		
2993.6	3.33E-04		
3794.9	3.03E-04		
<u>3905.9</u>	1.06E-03	<u>57</u>	-3.907000+6
		58	-3.999000+6
4180.2	9.08E-04	59	-4.032000+6
4354.8	6.05E-04	60	-4.378000+6
4449.3	6.05E-04	61	-4.549000+6
<u>4558.3</u>	6.05E-04	<u>62</u>	<u>-4.558000+6</u>
		63	-4.648000+6
		64	-4.683000+6
		65	-5.106000+6
		66	-5.366000+6

### Fluorine

E	ľ	E <sub>i</sub>	E <sub>γ</sub>	ľ	Eį	
197.1 (2) 1235.80 (10) 1261.1 (3) 1348.0 (5) 1356.5 (5) 1444.0 (4) 1458.4 (4) 1554.0 (6) 2352.6 (12) 2582.6 (2)	2700 (200) 100 20 (4) 120 (30) 265 (60) 15 (5) 33 (6) 8.0 (22) 1.0 (3) 20 (3)	197.1 1345.7 1458.4 1458.4 1553.9 1553.9 1458.4 1553.9 3906.2 2779.9	2653.8 (5) 2686.6 (5) 2993.6 (20) 3794.9 (15) 3905.9 (15) 4180.2 (15) 4354.8 (25) 4449.3 (25) 4558.3 (25)	5.4 (13) 3.7 (13) 1.1 (3) 1.0 (2) 3.5 (18) 3.0 (8) 2.0 (10) 2.0 (10) 2.0 (10)	3999.6 4032.5 3906.2 3906.2 4377.9 4552.4 4558.9 4558.9	

Attempts at modeling spectra not convincing Cascades/cross sections not matching up?

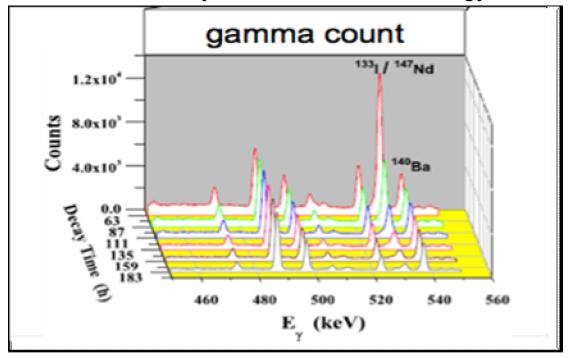
ENDF references ORNL measurements using NaI gamma spectrometers?

**Definitely Worth Further Evaluation** 

# Improved High Resolution Gamma Spectra of Fission Products-What Actinide?

State of the Art New Spectroscopy data: Time Dependent HPGe spectra from 15 High Yield Fission Products from

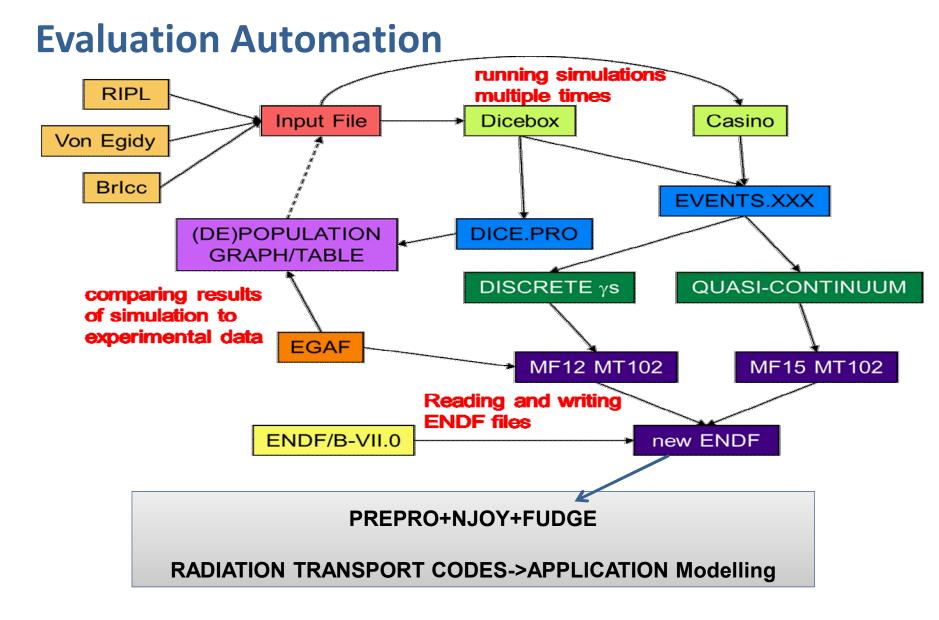
235U, 238U, 239Pu Fissioned by 5 narrow Neutron Energy Distributions



This New data should be in ENDF+233U, other Pu, 237Np

A. Tonchev, et. al.





Neil Summers, Brad Sleaford, LLNL



